



**QUEEN'S
UNIVERSITY
BELFAST**

Thermoforming carbon fibre-reinforced thermoplastic composites

McCool, R., Murphy, A., Wilson, R., Jiang, Z., Price, M., Butterfield, J., & Hornsby, P. (2012). Thermoforming carbon fibre-reinforced thermoplastic composites. *Proceedings of the Institution of Mechanical Engineers, Part L: Journal of Materials: Design and Applications*, 226(2), 91-102. <https://doi.org/10.1177/1464420712437318>

Published in:

Proceedings of the Institution of Mechanical Engineers, Part L: Journal of Materials: Design and Applications

Document Version:

Peer reviewed version

Queen's University Belfast - Research Portal:

[Link to publication record in Queen's University Belfast Research Portal](#)

Publisher rights

Copyright IMechE 2012

General rights

Copyright for the publications made accessible via the Queen's University Belfast Research Portal is retained by the author(s) and / or other copyright owners and it is a condition of accessing these publications that users recognise and abide by the legal requirements associated with these rights.

Take down policy

The Research Portal is Queen's institutional repository that provides access to Queen's research output. Every effort has been made to ensure that content in the Research Portal does not infringe any person's rights, or applicable UK laws. If you discover content in the Research Portal that you believe breaches copyright or violates any law, please contact openaccess@qub.ac.uk.

Thermoforming Carbon Fibre Reinforced Thermoplastic Composites

R. McCool, A. Murphy, R. Wilson, Z. Jiang, M. Price*, J. Butterfield and P. Hornsby

CEIAT, School of Mechanical and Aerospace Engineering, Queen's University Belfast,

Ashby Building, Belfast. N. Ireland, U.K. BT9 5AH.

Abstract

The use of carbon fibre composites is growing in many sectors but their use remains stronger in very high value industries such as aerospace where the demands of the application more easily justify the high energy input needed and the corresponding costs incurred. This energy and cost input is returned through gains over the whole life of the product, with for example, longer maintenance intervals for an aircraft and lower fuel burn. Thermoplastic composites however have a different energy and cost profile compared to traditional thermosets with notable differences in recyclability, but this profile for thermoplastics is not well quantified or documented. This study considers the key process control parameters and identifies an optimal window for processing thermoplastic composites, along with the effect this has on the final characteristics of the manufactured parts. Interactions between parameters and corresponding sensitivities are extracted from the results.

Keywords: Thermoforming, continuous fibre-reinforced thermoplastic, whole life modelling, composite material recycling-reuse.

*** Corresponding author:**

Tel.: +44 (0) 28 9097 4178; fax: +44 (0) 28 9066 1729.

E-mail address: m.price@qub.ac.uk (Mark Price).

1.0 Introduction

1.1 *Designing with new materials*

The introduction of new materials, particularly for aerospace products, is not a simple, quick or cheap task. New materials and associated processes require extensive and high cost qualification and must meet challenging manufacturing, strength, stiffness, durability, inspection and maintenance requirements, as well as emerging recycling-reuse targets, obligations and directives. Considering the structural design process, a design is traditionally conceived using experience in combination with highly idealized performance models. This is true for the simplest of components and for the complete vehicle structure. Given commercial pressures for reduced time to market and available data within early design, the ability to fully explore and assess the potential design space is highly constrained. Consequently designs and materials tend to be limited to the most feasible of a few considered, or limited to a known and previously used design and material combination. Moreover, in cases where new materials have been selected, the structural design may not initially maximise the material potential. A good example of this is the introduction of thermosetting composite structures in aerospace which were initially designed as if they were traditional metal materials with isotropic properties. Such design constraints can only be overcome with new understanding and the adoption of new design methods which account for the range of materials, and material to part processes available, as well as the complex interactions between the materials and the ultimate whole life structural performance.

In the case of composites this whole life aspect is fundamental in understanding the balance of energy and cost for a product. Each step in the manufacturing process consumes energy and incurs cost which can be relatively high for an individual process such as hand layup and autoclave moulding. However savings can be made with reduced assembly requirements and lower part counts if traditional DFMA principles can be successfully applied to products based on advanced materials. The exact combination of process and operational performance determines whether the savings outweigh the cost or vice versa. A recent study of the life-cycle performance of CFRP for buses and trucks by Song et al [1] showed that for those applications aluminium was a more cost effective replacement for steel than CFRP. The downfall of CFRP in this case was the high process costs and loss of value at the end of life. There were clearly substantial structural efficiency gains but these were not sufficient to make the case. This simply reinforces the old argument that performance gains cannot proceed in isolation from cost.

It is clear then, that to attain future vehicle structural efficiency levels, manufacturing cost reductions and a recyclable-reusable structure, it is necessary to know and understand relationships between the material, structural configuration and processing parameters, and the performance under the critical in-service environmental and loading conditions. It is also important that key attributes for recycling-reuse are known, measured and considered. Given the large number and range of parameters and the complexity of the potential relationships, this understanding must be built up in a systematic building block approach, which consecutively examines coupons, structural details, sub-components and full scale assemblies, developing the key interlinking relationships as the size, complexity and part count of the structure increases. In the past with such a scenario manufacturing and material design spaces as well as the structural configuration, have been constrained resulting in a simpler design space which enabled the generation of feasible designs. Slowly with time the individual manufacturing, configuration and material design spaces would expand and result in local and global performance improvements. This approach is not appropriate today as step changes are required with respect to improved development lead times, enhanced structural efficiency, reduced manufacturing cost and the requirement to develop more eco-aware structures. Even a single composite material configuration requires of the order of 500 coupon tests to determine properties before considering structural details or assemblies. Certification of a major structure can require the order of 10^4 tests. Moreover, such targeted improvements require design tools which can rapidly assess new materials and processes, highlighting the need to further develop computational modelling and reduce dependence on experimental methods in product development. Further, material suppliers provide limited physical properties or processing values which seldom go beyond their own requirements for quality control purposes. Designers and end users need a range of values, i.e. a processing window, which they can use to tailor material processing parameters for a given processing method in order to optimise their overall system. This requires the determination of final characteristics for the variation across a range of thermal and physical parameters.

This wider systems view requires a building block approach, hence this article documents the initial coupon level development of a continuous fibre-reinforced thermoplastic laminate thermoforming process, identifying the key interactions between processing parameters and resulting part performance characteristics. This is a broad and challenging problem and in this work an experimental design approach is necessarily adopted as a starting point.

Therefore this work aims to:

1. Select an initial set of parameters which are relevant for manufacturing, service and recycling.
2. Obtain values for the key manufacturing parameters which define an optimal processing window.
3. Critically assess the results and specify a baseline for advanced manufacturing trials.

In the next section the relevant materials and processes used herein are introduced before describing how these and the appropriate performance characteristics are chosen. Section three then presents preliminary material analysis and the methodology used to identify the key interactions between the coupon thermoforming parameters and the resulting performance characteristics. The measurement techniques used and the resulting experimental data are presented in sections four and five. Section six concludes the article with a summary of the key findings and the processing window obtained.

2.0 Material and process background

In order to consider which parameters are relevant it is necessary to review some background on both the material itself and the manufacturing processes.

2.1 Continuous fibre-reinforced thermoplastic laminates

The majority of composite materials used today in aerospace products are continuous fibre-reinforced thermosets. However, since their introduction in the 1980's, continuous fibre-reinforced thermoplastic materials have matured with a range of matrix materials such as Polyetherimide (PEI)[2], Polyether ether ketone (PEEK)[3], Polyethersulphone (PES)[4] and Polyphenylene sulfide (PPS)[5] used in combination with carbon fibre. What is more, there have been a number of examples of such materials being used on commercial transport aircraft, including fixed wing leading edges[6], wing-nacelle pylon panels[7] and wing flap ribs[7] proving the material has promise. However these are bespoke applications and there have followed no general rules or publically available data characterising their behaviour in a wide range of conditions.

A thermoplastic matrix has a number of potential advantages over a thermosetting resin. Thermoplastic materials typically exhibit high toughness, high service temperature and shorter manufacturing cycle times, as part manufacture does not rely on a chemical process. In

addition, thermoplastic raw materials do not have short shelf lives nor do they require refrigerated storage as with typical aerospace thermosetting resins. Moreover, thermoplastic resins are re-processable, making it possible to efficiently repair local defects in service, an advantage for large, expensive unitised structures. This potential for reprocessing also has a significant advantage when it comes to recycling and reuse of structures at the end of the vehicle's life. Of course they have a number of disadvantages too, including higher processing temperatures (e.g. Epoxy 150°C[8] and PEEK 380°C[9]), higher raw material costs and currently a lack of available design and manufacturing data.

Compared to component manufacture using thermosetting prepregs, use of thermoplastic laminates as a raw material, introduces the potential for faster final part production. Typically the raw materials for thermoplastic laminate consist of pre-consolidated layers of semipregs, with the supplied fabric plies impregnated by thermoplastic resin films in a preproduction manufacturing stage. It is supplied as a flat sheet. Although higher levels of raw material processing results in a corresponding higher cost, the pre-consolidated laminate sheets offer production benefits as they can be readily processed using a number of derivatives of the thermoforming family of processes, such as matched die forming, rubber thermoforming, diaphragm forming etc. Eliminating the need for layup processes in the production environment, also improves quality levels and reduces the potential for variability in finished pieces. Examples of processes currently under investigation which have the potential to be used in conjunction with thermoforming for the manufacture of complex components include thermofolding [10] and thermoplastic welding [10, 11].

In this work PPS was chosen for its material properties and for its emergence as a leading thermoplastic matrix material for aerospace components[7]. The chemical structure of PPS consists of para-phenylene units alternating with sulphide linkages, such an arrangement leads to a very rigid macromolecular chain, which exhibits superior physical and mechanical properties[12]. PPS is very inert and exhibits excellent chemical resistance including resistance to typical aircraft fuel and hydraulic fluids [12]. Another important characteristic of PPS is its "thermosetting-thermoplastic" behaviour. The potential for material chemical reactions including cross-linking of molecular chains (chain extension, oxidative cross-linking, and thermal cross-linking) occurring around and above the material melt temperature [13, 14], results in good dimensional stability at elevated service temperatures, but these material behaviours also result in reduced potential for reprocessing and thus recycling, because they are irreversible.

2.2 Thermoforming fibre reinforced thermoplastics

Thermoforming [10, 15-18] is a process which is long established in the manufacture of thin walled plastic containers and has enabled the manufacture of commercial products such as yoghurt pots and blister packs. It enables rapid transformation of semi-finished raw material into the required design form by the combined action of heat and pressure. Relatively short forming times are made possible by the fast processing potential of thermoplastics [10]. The process can be characterized in terms of the physical shape change and thermal history of the blank during the complete process shown in Figure 1.

The first stage involves heating the thermoplastic, pre-consolidated laminate to a temperature above the melt temperature of the thermoplastic matrix system, as shown in Figure 1. De-consolidation occurs in the raw material blank during the heating phase, resulting from a release in the stored elastic energy in the form of residual stress which originates from the initial raw material compaction process [19]. The melt temperature of pure PPS is around 280°C [5]. Melt phase thermoforming permits intra-ply and inter-ply slip, and at these elevated temperatures the dominant mode of deformation is slip parallel to the individual plies [16]. Therefore, process temperature determination is of great importance because without sufficient melting of the thermoplastic matrix, forming can result in matrix cracking, fibre buckling, fibre bridging, wrinkling and therefore poor part quality and strength performance, especially in areas of sharp contours and geometric detail. However, thermoplastic matrix material is also sensitive to temperature history, influencing induced residual stress, crystallinity and thus mechanical performance of the final part. Moreover, as the thermoforming process requires blank temperatures above the matrix melt temperature, chemical reactions (cross-linking of molecular chains) can occur, and as noted previously can impact in-service performance and reprocessing.

In the second stage, the material blank is indexed into the forming station (Figure 1) at a predefined temperature, usually just above the forming temperature to take into account thermal heat losses during indexing between stations. Once located in the forming station the blank material is consolidated into a target shape by the closure of the matched die moulds. The role of the mould is twofold. Firstly the mould re-consolidates the material plies and gives the finished product its shape, thickness and surface finish. Secondly, the mould temperature is set at a predefined temperature to allow controlled part cooling and therefore solidification of the part prior to part extraction from the mould, Figure 1. The final part thickness is governed by a combination of factors including mould tool geometry, ply layup, matrix chemical reactions and shrinkage. The mould tool design has a significant influence on part quality, including thickness

variation as well as local and global part fibre volume fraction. Also, the level of tool compaction of the deconsolidated raw material blank has the potential to significantly influence the inter-laminar shear strength of the part, if voids in the form of entrapped air, moisture and volatiles are not eliminated during the reconsolidation.

The cooling rate after forming, normally determined by the blank temperature (x) and mould tool temperature (y), is potentially one of the most important processing factors, significantly influencing matrix material crystallinity and residual stress [20]. For semi-crystalline thermoplastics, a higher cooling rate will tend to result in a lower peak crystallisation temperature and lower crystallinity levels [20], and less matrix shrinkage. A high level of crystallinity will result in higher levels of static strength mechanical properties and chemical resistance, but lower matrix fracture toughness properties. In addition, the mould tool temperature will influence the cycle time (z) of the process as processed parts can only be extracted from the mould when they are structurally rigid and sufficiently cooled so that physical handling is possible and safe. Finally, x, y & z all significantly impact the energy footprint of the process.

3.0 Methodology

The preceding description indicates that there are several processing parameters which can have an influence on final performance, but three of these initially appear to characterise the forming process and potentially have an influence on the performance of the formed parts. The process parameters identified are:

1. the raw material blank forming temperature,
2. the mould tool temperature
3. the mould tool consolidation (compaction metric).

In this study the mould tool consolidation metric will be simplified into a 1D metric, describing the ratio of the nominal closed tool thickness over the nominal raw material thickness. Herein this ratio will represent the tool design and is labelled as the mould tool consolidation compaction ratio.

To study any potential non-linear process effects, the three process parameters will be examined at three magnitudes, a high level, a medium level and a low level.

3.1 Manufacturing Process Parameters

In order to identify the magnitudes of the parameters for the study a series of preliminary processing trials were carried out to establish the approximate limits for each. A Dynamic Mechanical Thermal Analysis (DMTA) was carried out to establish the temperature range for which thermoforming is possible, and simple trials heating and cooling samples helped to identify de-consolidation and re-consolidation times. Samples were heated for varying times and rapidly cooled to freeze the deconsolidated state so that the appropriate heating times for a range of temperatures could be established. These identified that a heating time of three minutes was sufficient, and this was then used for all specimens. The trials also identified blank and mould temperatures and compaction ratios as described in the following three sections.

3.1.1 Blank Forming temperature window

Analysis of the DMTA data from the preliminary trials shows that the melting temperature (T_{melt}) of the as-supplied material was approximately 285 °C, as shown in Figure 2. However, the data shows that the material still exhibits a considerable modulus of 3.77 GPa at this temperature, which indicates that the material is too stiff for thermoforming. Thermoforming is only viable at temperatures where the modulus would tend to zero in a DMTA analysis. In this case it was found to be well above the melt temperature with the minimum level raw material blank forming temperature being defined as 310 °C. To establish the effect of elevated forming temperatures on the physical and mechanical properties of the formed parts, two further temperature levels of 340 °C and 370 °C were selected.

3.1.2 Mould temperature window

The crystallisation kinetics of PPS are mainly influenced by the cooling rate of the process which is dictated by the raw material blank forming temperature ($T_{forming}$) and the mould tool temperature (T_{mould}). To understand and quantify the relationships, the mould tool temperature will be experimentally examined at 50 °C, 110 °C and 170 °C, again, based on simple explorative forming trials.

3.1.3 Mould tool consolidation window

Pre-consolidated laminates are supplied with a thickness which is dependent on ply layup and fibre volume fraction. The raw material laminate used in this work has an average thickness of 1.80 mm, with maximum and minimum recorded raw material thickness values of 1.87 mm and

1.68 mm respectively. Preliminary re-consolidation trials were carried out to determine the range of consolidation compaction ratios (ratio of the nominal closed tool thickness over the nominal raw material thickness), with cross sectional images used to assess part consolidation and void content. Three consolidation compaction ratios (CCR) of 1.00, 0.97 and 0.94 were determined for examination within the manufacturing trials. These compaction ratios were selected to represent potential limits for the final part thickness achievable during the forming process from the initial variability in the raw material thickness. These compaction ratios represent closed tool thicknesses of 1.80, 1.75 and 1.70 mm respectively.

3.1.4 Parameter Values for the experiments

Thus considering three processing parameters at three levels requires a set of manufacturing trials which follows a Taguchi three factor, three level orthogonal array, L9 [21-22]. Using the fractional factorial experimental design method in the present case, only nine manufacturing trials are required to identify the dominant factors and their potential interactions.

3.2 Performance Parameters

To identify interactions between processing parameters and performance characteristics each coupon set produced within the manufacturing trial was subsequently prepared for a range of post processing analysis and test. To define the relevant performance parameters an exemplar application is required. The manufactured coupons represent the local design (laminare stacking sequence and thickness) of an outboard wing rib vertical reinforcing strut, designed to control rib web buckling under compression and shear loading. The thickness of the represented structure is 1.8 mm and consists of a six ply, five harness satin woven carbon fibre (T300 JB) reinforced PPS pre-consolidated laminate with a stacking sequence of $[(0,90)_3]_s$, and a fibre volume content of $50 \pm 3\%$. Table 1 presents the supplied material properties of the laminate and for the neat PPS matrix. There are many characteristics which define the performance of a part throughout its life, from manufacture to disposal. To initiate the framework this study was limited to a set of seven key characteristics covering processing quality and cost, in-service strength performance, and end-of-life recyclability:

- 1) Variation of Fibre Volume Fraction of the as-processed coupons,
- 2) Variation of surface roughness of the as-processed coupons,
- 3) Derived production cycle time of the employed process parameters,
- 4) Flexural strength of the as-processed coupons,
- 5) Degree of crystallinity of the as-processed coupons,
- 6) Cold crystallisation temperature of the as-processed coupons, and finally

7) Reprocessing melt temperature of the as-processed coupons,

The measurement methods and rationale for selection of each of these will be expanded below. For each test type, specimens were water-cut from the manufacturing trial coupons and prepared as required for the individual tests.

3.2.1 Volume fraction characteristics

Fibre volume fraction (V_f) is a measure of fibre content relative to the total material make up and is thus an important indicator of process quality. Correct thermoforming of the raw material will produce close tolerance part fibre volume fraction, equal to the original raw material fraction. Specimens were weighed in both air and methanol using a precision scale (Sartorius LA620P) and the density buoyancy method used to approximately determine the fibre volume fraction, assuming no voids and the individual density of the matrix and the fibre constituent materials. Although void content has not been taken into account when examining fibre volume fraction for this work, Wakeman's findings [23] indicate that pressure forming methods can be used effectively to control void content minimizing their influence on finished part form properties such as fibre volume fraction. Indicative figures for an equivalent material system using stamp forming methods, show that void content in 3mm thick sheets of carbon fibre reinforced PA66 started at 1.5% in the pre-consolidated state, rising to 15% during heating but finishing at 1% after forming[23].

3.2.2 Surface roughness characteristics

Surface roughness (R_a) provides a measure of surface finish and ultimately final product quality. The finish derived using given thermoplastic based material and process combinations influences downstream finishing activities such as surface pre-treatment, priming and painting processes which are in turn, used to meet the joining and customer finish requirements for the part. As the same mould surfaces were used for all manufacturing trials (ground nickel plated carbon steel platens) the influence of the process parameters can be appraised. Surface roughness, measured in μm , is quantified here by vertical deflections of a stylus as it is traced across the specimen surface, herein using a Taylor-Hobson Talysurf.

3.2.3 Derived production cycle time

Process cycle time is a key indicator of manufacturing recurring cost, and combined with temperature data characterises the process energy footprint. It is particularly relevant at coupon level analysis where other associated key recurring costs such as raw material, pre and post process setup and inspection times are necessarily constant.

The derived production cycle times were determined by interrogating the individual temperature data for the trial specimens and identifying the additional dwell times due to the fixed blank heating and mould cooling times, removing these to calculate the optimised blank heat up and blank cool down times. The derived production cycle time t (seconds) was then calculated for each trial based on the minimum time taken for thermal de-consolidation (t_a), re-consolidation (t_b), natural cooling in the mould (t_c), and ambient part cooling (t_d) after mould ejection, to a safe handling temperature (30°C in this case), and the summation of the individual times thus:

$$t = t_a + t_b + t_c + t_d \quad (1)$$

3.2.4 Flexural strength characteristics

Maximum flexural strength (σ_f) is an important indicator of the in-service static strength of the formed part and an indicator on the quality of reconsolidation during forming. The test coupons were based on the ASTM D790 standard.

3.2.5 Crystallinity and melt temperature characteristics

Differential Scanning Calorimetry (DSC) was used to determine the degree of crystallinity, cold crystallisation temperature and reprocessing melt temperature of the processed manufacturing trial coupons. The degree of crystallinity is measured as it is an important indicator of material static strength and fracture toughness, and also the level of irreversible cross-linking chemical reaction which has occurred during processing. The cold crystallisation temperature is an important metric with respect to the in-service temperature ceiling, although it is not the ultimate temperature limit of the matrix material, service above this temperature will alter the crystallinity and thus part behaviour. The reprocessing melt temperature is also measured, indicating the level of irreversible chemical reaction during processing.

The properties were measured following the ISO 11357 standard with a 10 mg specimen over a temperature range of 20 to 310 °C, at a heating rate of 20 °C per minute. Repeat measurements were carried out for each manufacturing trial coupon, and the mean values calculated. The area

of the melting peak and cold crystallisation peak (ΔH) in the specific heat flow versus specimen temperature curves, were determined to calculate the degree of crystallinity (χ), in accordance with Equation 2:

$$\chi = \frac{\Delta H}{(1 - W_f) \Delta H_f} \quad (2)$$

where W_f is the fibre weight fraction of the processed material, and ΔH_f represents the enthalpy of fusion for 100% crystalline PPS, taken as 112 J/g [24].

It is important to note that the final degree of crystallinity (χ_{final}) does not represent the crystalline level of the processed coupon, since the samples experience a re-crystallisation during the DSC characterisation. Thus the cold crystallinity (χ_{cold}) i.e. that measured at 20 °C should be subtracted from the final measured crystallinity. Therefore, the degree of crystallinity for the as-processed coupons is calculated using Equation 3:

$$\chi_c = \chi_{\text{final}} - \chi_{\text{cold}} \quad (3)$$

3.3 Test Procedures

The thermoforming trials were carried out using a ten ton laboratory platen press. The press consists of precision ground nickel plated carbon steel platens with independent platen temperature control. The platen press enabled flat plate thermoforming with constant consolidated thickness. In addition to temperature control, coupon thickness control was maintained by incorporating precision ground steel shims of known thickness between the platens. Raw material laminate coupons with major length and width dimensions of 110 mm and 20 mm respectively were water-jet cut from a 480 mm x 480 mm raw material plate. Prior to thermoforming, the test pieces were mounted on a transport fixture and then introduced into the heating chamber. Temperature mapping was achieved using an eight channel K-type thermocouple temperature logging device. The test pieces were rapidly raised to a maximum forming temperature ($T_{\text{forming max}}$) i.e. the $T_{\text{forming}} + 10^\circ\text{C}$ to take into account heat losses due to transport between heating environment and the forming station, at which point they were manually indexed into the forming position. Once in place the heated test pieces were consolidated within the press, and after three minutes the samples were ejected from the tool into the ambient environment. The magnitude of the three process parameters for the nine fractional factorial manufacturing trials are summarised in Table 2.

4.0 Results and discussion

4.1 Global results analysis

Results from the characteristic experiments are summarised in Table 3. The first step is to conduct an Analysis of Means (ANOM) where the average performance for each factor level is computed and plotted. These plots allow an immediate visual inspection of the influence of the process parameters, Figure 3. Moreover, using the ANOM data, a series of virtual optimisations can be performed, Table 4. The optimum objective for fibre volume and surface roughness variation, and derived production cycle time is a minimum value. For in-service flexural strength, the degree of crystallinity and reprocessing melt temperature the optimisation objective is for a maximum value. It is also possible to predict the value of the characteristic for the optimised process parameters (right hand column of Table 4). There is a clear indication that to achieve desirable values in the majority of the characteristics, the material should be processed using a raw material blank forming temperature of 310 °C and a mould tool temperature of 170 °C.

The ANOVA of the experimental data also enables the comparison of the relative contribution of each process parameter to the characteristic properties. Table 5 shows the significant factors. Examining the pooled analysis calculated at 95% confidence limits, the raw material blank forming temperature is the dominant factor influencing the surface roughness variation and reprocessing melt temperature. Whereas the mould tool temperature is the main factor affecting the derived production cycle time, in-service flexural strength, degree of crystallinity, and the cold crystallisation temperature.

4.2 Detailed analysis of process characteristics

Examining the measured variation in fibre volume fraction of the manufacturing trial coupons, Table 3, no significant variation is seen within the results, indicating that across the complete processing window fibre volume fraction is not significantly modified, which demonstrates that the study has been conducted within a practical processing window. Thus, there is no single dominant process parameter determined which is controlling fibre volume tolerance.

Table 3 also presents the variation of surface roughness with the data indicating the importance of the forming temperature but also the generally large magnitude of variation across the

complete series. This variation can potentially be attributed to the spatial nature of the composite material and the numerous random measurement locations on the laminate surfaces.

4.3 Detailed analysis of in-service characteristics

Examining the results in Figure 6 and Table 3 there is a clear trend linking the flexural stress and the mould tool temperature, with higher mould tool temperatures resulting in higher flexural stress. Figure 5 shows a strong positive correlation between flexural strength and crystallinity, and from Table 5 it can be seen that the degree of crystallinity is strongly linked to the mould tool temperature.

Again examining Figure 4 the effect of blank forming temperature can be observed. Higher blank forming temperatures produce lower flexural stress. This suggests that the increased blank forming temperature increases the level of matrix chemical reactions during processing (cross-linking of molecular chains) and this reduces the potential for crystallisation and the associated improvement in matrix static mechanical strength.

The DSC plots for the manufacturing trials are shown in Figure 6 along with an additional DSC plot for an unprocessed raw material sample (labelled unprocessed). Peaks related to cold crystallisation are clearly visible on the curves for the processed sample, but not on the unprocessed sample. Two peak temperatures of cold crystallisation are found around 113 and 134°C respectively, depending on the mould tool temperature, with higher mould tool temperatures resulting in lower cold crystallisation temperatures. Such a trend can be ascribed to the degree of alignment of molecular chains, which can occur with greater ease at higher mould temperatures. For well aligned molecular chains, cold crystallisation is prone to occur with less thermal excitation, i.e. at a lower temperature.

4.4 Recyclability characteristics

The variation in processed material melt temperature measured for all manufacturing trials was only 6 °C, Table 3, indicating a modest variation in the level of matrix chemical reactions across the complete processing window examined. It has been reported that several forms of chemical reactions can occur simultaneously when PPS is processed in air at temperatures between the melting point and 400°C. The reactions include chain extension, oxidative cross-linking, and thermal cross-linking [14]. As shown in Figure 4, a higher forming temperature results in a

decrease in the measured degree of crystallinity, which is indicative of a higher level of cross-linking chemical reactions under such processing conditions. Thus higher blank forming temperatures are generally accompanied by an increase in cross-linking chemical reactions, and a reduction in the potential for reprocessing. However, the reduced potential for reprocessing is modest for the complete processing window examined herein.

5.0 Conclusions

This article documents the coupon level development of a continuous fibre-reinforced thermoplastic laminate thermoforming process, identifying the key interactions between processing and a select number of performance characteristics. For the examined material and process, the undertaken experimental and statistical analysis has indicated that to achieve desirable values in the majority of the characteristics, the material should be processed using a raw material blank forming temperature of 310°C and a mould tool temperature of 170°C. In addition, for the range of process parameters and material examined, the experimental analysis has established that the raw material blank forming temperature is the dominant factor influencing the surface roughness variation and reprocessing melt temperature. Whereas the mould tool temperature is the main factor affecting the derived production cycle time, in-service flexural strength, degree of crystallinity, and the cold crystallisation temperature of the processed coupons.

A fractional factorial experiment design method to identify interactions between processing parameters and resulting part performance characteristics, has been demonstrated as appropriate and efficient for initial coupon level analysis.

Acknowledgements

The authors would like to thank the Department of Employment and Learning (Northern Ireland) and Queens University for their generous support of this project ['Sustainable Transport – Strengthening the All-Island Research Base' Programme].

References

- [1] Song Y.S, Youn J.R., Gutowski T.G. "Life Cycle Energy Analysis of Fibre reinforced composites", Composites Part A, Vol 40, pp1257-1265, 2009.

- [2] Bersee HEN, Beukers A. Consolidation of Thermoplastic Composites. *Journal Of Thermoplastic Composite Materials* 2003;16(5):433-455.
- [3] Ye L, Friedrich K, Cutolo D, Savadori A. Manufacture of CF/PEEK composites from powder/sheath fibre preforms. *Composites Manufacturing* 1994;5(1):41-50.
- [4] Yumitori S, Wang D, Jones FR. The role of sizing resins in carbon fibre-reinforced polyethersulfone (PES). *Composites* 1994;25(7):698-705.
- [5] Ye L, Chen Z-R, Lu M, Hou M. De-consolidation and re-consolidation in CF/PPS thermoplastic matrix composites. *Composites Part A-Applied Science And Manufacturing* 2005;36(7):915-922.
- [6] Mitschang P, Blinzler M, Wöginger A. Processing technologies for continuous fibre reinforced thermoplastics with novel polymer blends. *Composites Science and Technology* 2003;63(14):2099-2110.
- [7] Díaz J, Rubio L. Developments to manufacture structural aeronautical parts in carbon fibre reinforced thermoplastic materials. *Journal of Materials Processing Technology* 2003;143-144:342-346.
- [8] Sohn M-S, Hu X-Z. Processing of carbon-fibre/epoxy composites with cost-effective interlaminar reinforcement. *Composites Science and Technology* 1998;58(2):211-220.
- [9] Jar PYB, Mulone R, Davies P, Kausch HH. A study of the effect of forming temperature on the mechanical behaviour of carbon-fibre/peek composites. *Composites Science and Technology* 1993;46(1):7-19.
- [10] Offringa AR. Thermoplastic composites--rapid processing applications. *Composites Part A: Applied Science and Manufacturing* 1996;27(4):329-336.
- [11] Ahmed TJ, Stavrov D, Bersee HEN, Beukers A. Induction welding of thermoplastic composites--an overview. *Composites Part A: Applied Science and Manufacturing* 2006;37(10):1638-1651.
- [12] Béland S. High performance thermoplastic resins and their composites. Park Ridge: Noyes Publications, 1990.
- [13] Brady DG. The crystallinity of poly(phenylene sulfide) and its effect on polymer properties. *Journal of Applied Polymer Science* 1976;20(9):2541-2551.
- [14] Hawkins RT. Chemistry of the cure of poly (p-phenylene sulfide). *Macromolecules* 1976;9(2):189-194.

- [15] Scherer R, Friedrich K. Inter- and intraply-slip flow processes during thermoforming of cf/pp-laminates. *Composites Manufacturing* 1991;2(2):92-96.
- [16] Schuster J, Friedrich K. Modeling of the mechanical properties of discontinuous-aligned-fiber composites after thermoforming. *Composites Science and Technology* 1997;57(4):405-413.
- [17] Walczyk DF, Yoo S. Design and fabrication of a laminated thermoforming tool with enhanced features. *Journal of Manufacturing Processes* 2009;11(1):8-18.
- [18] Hou M, Ye L, Mai YW. Manufacturing process and mechanical properties of thermoplastic composite components. *Journal of Materials Processing Technology* 1997;63(1-3):334-338.
- [19] Gutowski TG. A resin flow/fiber deformation model for composites. *Sampe Q-Soc Adv Mater Process Eng* 1985;16(4):58-64.
- [20] Parlevliet PP, Bersee HEN, Beukers A. Residual stresses in thermoplastic composites--A study of the literature--Part I: Formation of residual stresses. *Composites Part A: Applied Science and Manufacturing* 2006;37(11):1847-1857.
- [21] Box GE, Hunter WG, Hunter JS, Hunter WG. *Statistics for Experimenters: Design, Innovation, and Discovery*. Wiley, 2005.
- [22] Yuin W, Wu A. *Taguchi Methods for robust design*. ASME, 2000.
- [23] M.D. Wakeman, P. Blanchard, J.-A. E. Manson. VOID EVOLUTION DURING STAMP-FORMING OF THERMOPLASTIC COMPOSITES. 15th International Conference on Composite Materials (ICCM-15), Durban, South Africa, 27 June to 01 July 2005.
- [24] Cebe P. Review of recent developments in polyphenylene sulfide). *Polymers & Polymer Composites* 1995;3(4):239-266.

Table 1. CF/PPS laminate properties and Neat PPS matrix properties.

Fibre Areal Weight (g/m ²)	Weight Ratio Warp:Weft	Resin Content by Volume (%)	PPS Density (kg/m ³)	PPS Melt Temperature (°C)	PPS Glass Transition Temperature (°C)
285	50:50	50	1350	280	90

Table 2. L9 array with factors and levels.

Trial	Blank forming temperature (°C)	Mould tool temperature (°C)	Consolidation compaction ratio
1	310	50	0.94
2	310	110	0.97
3	310	170	1.0
4	340	50	0.97
5	340	110	1.0
6	340	170	0.94
7	370	50	1.0
8	370	110	0.94
9	370	170	0.97

Table 3. Table showing DOE results.

Trial	Variation of Fibre Volume Fraction	Variation of surface roughness	Derived production cycle time	Flexural strength	Degree of crystallinity	Cold crystallization temperature	Reprocessing melt temperature
	($\Delta\%$)	($\Delta\mu\text{m}$)	(sec)	(MPa)	(%)	($^{\circ}\text{C}$)	($^{\circ}\text{C}$)
1	8.15	1.14	430	739	12.6	135	278
2	5.39	1.05	522	765	20.8	132	278
3	5.9	0.96	590	1015	29.4	113	280
4	7.2	1.56	458	679	11.4	137	277
5	7.07	1.58	533	721	12.5	137	279
6	4.49	1.48	604	967	20.9	114	277
7	5.11	1.16	483	645	8.9	134	275
8	4.1	1.49	554	756	14.1	138	274
9	5.81	1.18	624	955	20.8	114	274

Table 4. Optimised process parameters with statistically predicted values.

		Blank forming temperature	Mould tool temperature	Consolidation compaction ratio	Statistical Prediction
Output Optimized	Target	(°C)	(°C)		
Variation of Fibre Volume Fraction ($\Delta\%$)	Min	310	50	0.97	4.5 \pm 3.2
Variation of surface roughness ($\Delta\mu\text{m}$)	Min	370	170	1.00	1.4 \pm 0.3*
Derived production cycle time (sec)	Min	310	50	0.94	438 \pm 14.9
Flexural strength (MPa)	Max	310	170	0.94	979 \pm 87
Degree of crystallinity (%)	Max	310	170	0.97	27.8 \pm 5.3
Cold crystallization temperature (°C)	Max	310	170	0.97	136 \pm 5
Reprocessing melt temperature (°C)	Max	310	170	1.00	280 \pm 1
(significant factors only, 95% confidence limits, * = insignificant)					

Table 5. Manufacture trial ANOVA results.

Unpooled (system error included)								
		Characteristics						
		Variation of Fibre Volume Fraction (%)	Variation of surface roughness ($\Delta\mu\text{m}$)	Derived production cycle time (sec)	Flexural strength (MPa)	Degree of crystallinity (%)	Cold crystallization temperature ($^{\circ}\text{C}$)	Reprocessing melt temperature ($^{\circ}\text{C}$)
Process parameters	Blank forming temperature ($^{\circ}\text{C}$)	27%	80%	7%	4%	22%	1%	81%
	Mould tool temperature ($^{\circ}\text{C}$)	26%	9%	93%	95%	72%	97%	0%
	Consolidation compaction ratio	4%	7%	0%	1%	1%	0%	16%

Pooled 95% Confidence (* = insignificant)								
		Characteristics						
		Variation of Fibre Volume Fraction (%)	Variation of surface roughness ($\Delta\mu\text{m}$)	Derived production cycle time (sec)	Flexural strength (MPa)	Degree of crystallinity (%)	Cold crystallization temperature ($^{\circ}\text{C}$)	Reprocessing melt temperature ($^{\circ}\text{C}$)
Process parameters	Blank forming temperature ($^{\circ}\text{C}$)	*	73%	6%	*	19%	*	80%
	Mould tool temperature ($^{\circ}\text{C}$)	*	*	93%	93%	70%	96%	*
	Consolidation compaction ratio	*	*	*	*	*	*	15%

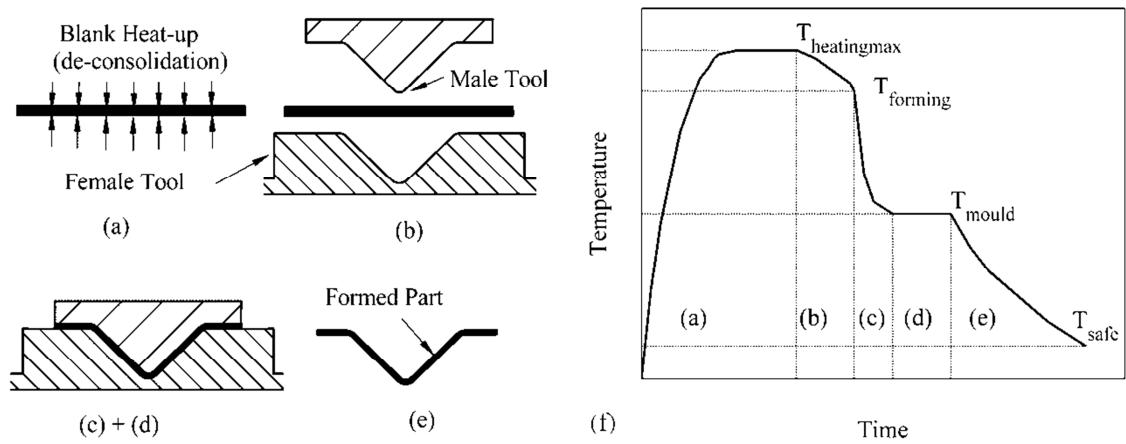


Figure 1. Key stages in the thermoforming process for reinforced thermoplastic composites, including thermal history.

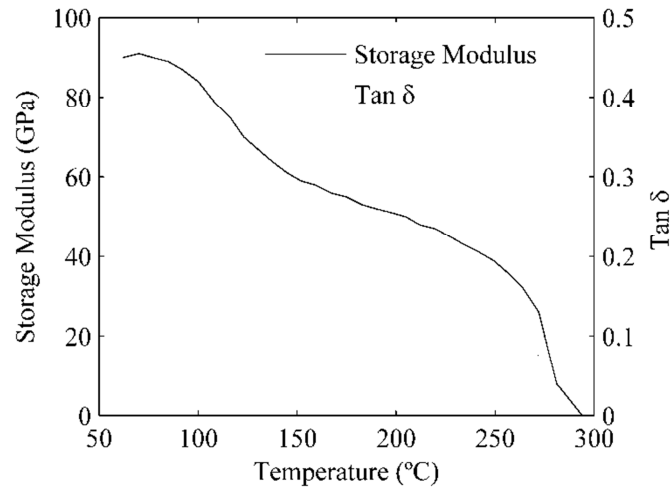


Figure 2. Raw material CF/PPS laminate - Storage modulus and damping factor $\tan \delta$ versus temperature obtained by DMTA.

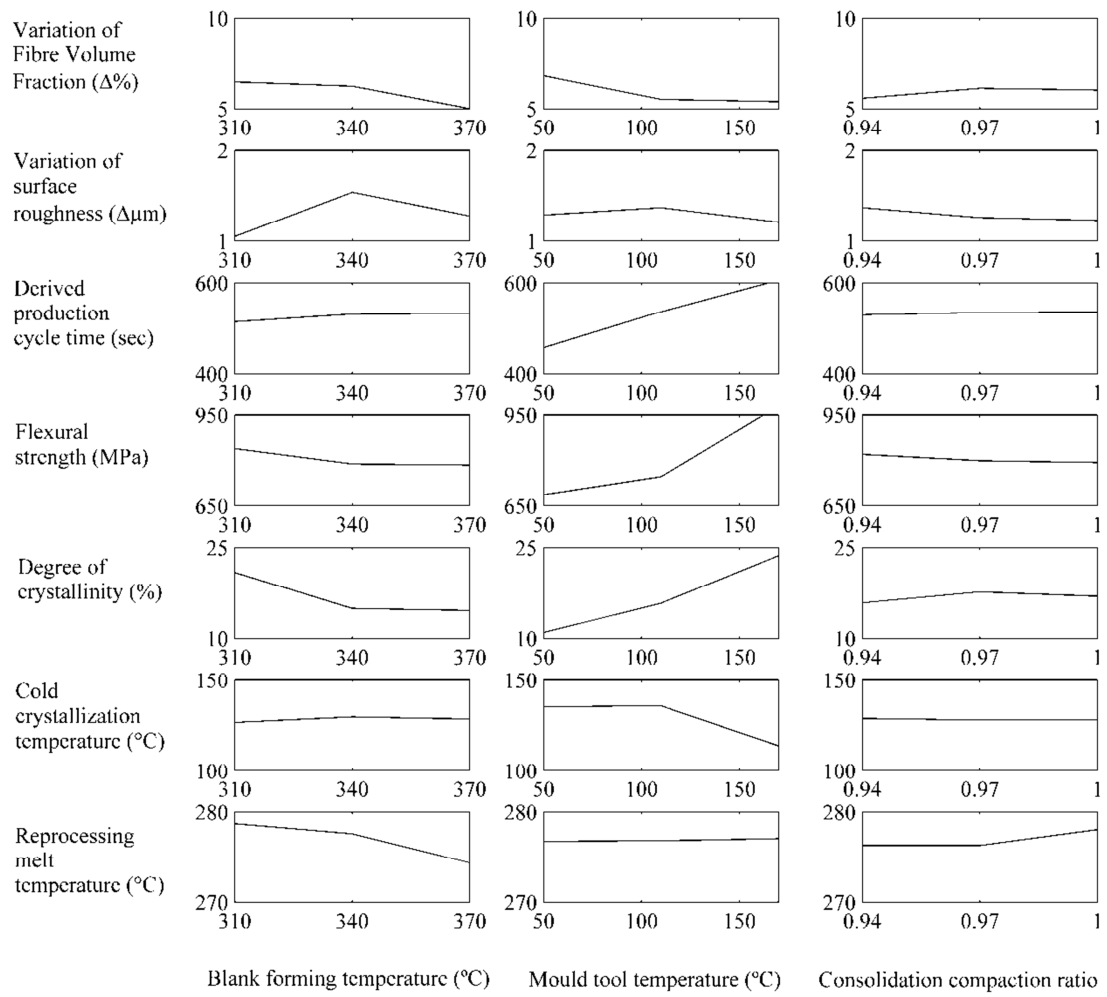


Figure 3. Manufacturing trial main effects plots for all outputs.

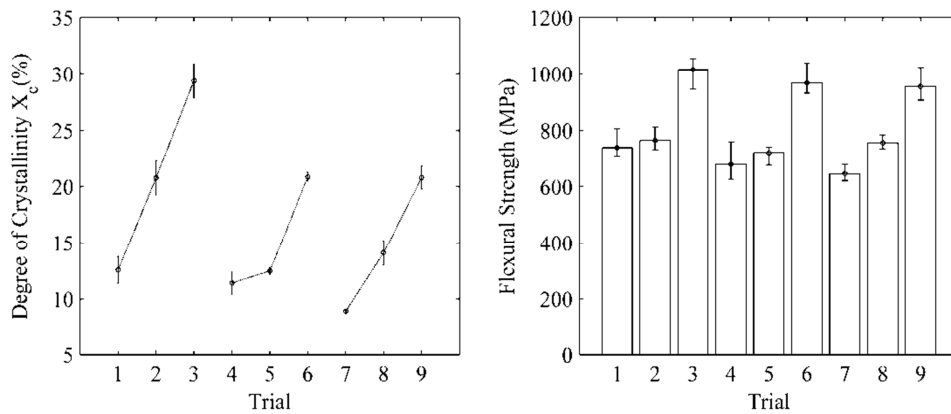


Figure 4. Degree of Crystallinity X_c and Flexural Strength σ_f processed CF/PPS laminates.

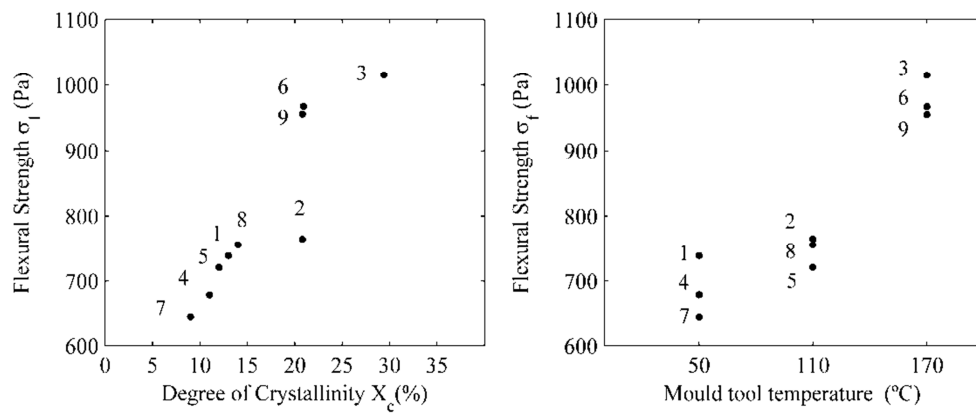


Figure 5. Relationship between Degree of Crystallinity X_c , mould tool temperature and Flexural Strength σ_f .

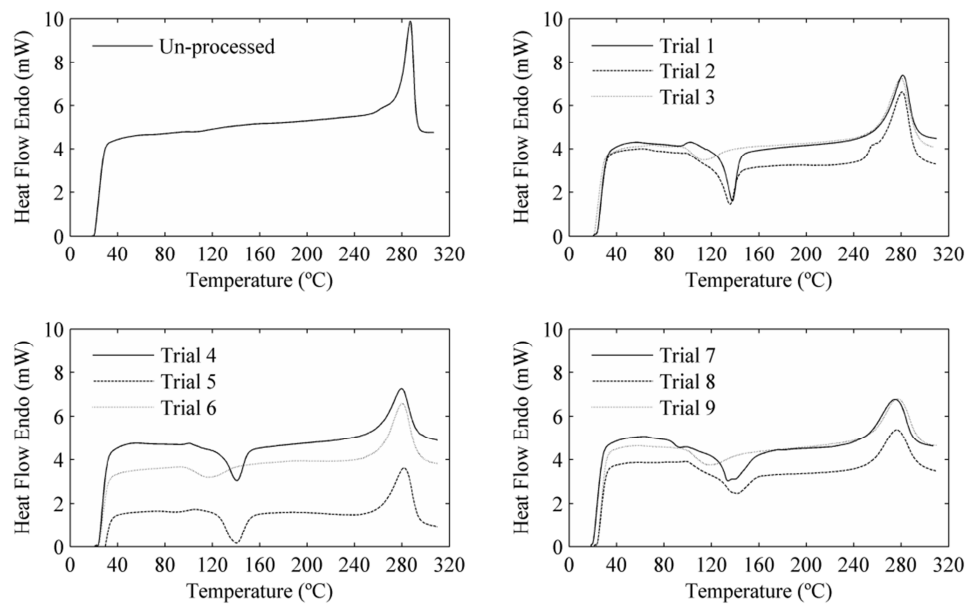


Figure 6. DSC plots for all trials and un-processed material.